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(54) [Title of the Invention]

Resin Composition for Calender Molding

(57) [Summary]

[Object] To provide a composition for calender molding that is composed of an amorphous polyester resin and can be molded into sheeting by means of calender molding.

[Means of Achievement] Obtaining a resin composition that contains as essential components a fatty acid ester-based montanic acid wax and an amorphous polyester in which a portion of the

ethylene glycol component among the terephthalic acid and ethylene glycol components is substituted by 1,4-cyclohexanedimethanol.

**[Claims]**

**[Claim 1]** A resin composition in which an amorphous polyester resin and a fatty acid ester-based montanic acid wax are included as essential components, wherein the resin composition for calender molding comprises a copolymer in which a portion of the ethylene glycol component among the terephthalic acid and ethylene glycol components of the amorphous polyester resin is substituted by 1,4-cyclohexanedimethanol.

**[Claim 2]** The resin composition for calender molding according to Claim 1, wherein the blending ratio of the amorphous polyester resin and fatty acid ester-based montanic acid wax is 0.1 to 5.0 weight parts montanic acid wax per 100 weight parts of the amorphous polyester resin.

**[Detailed Description of the Invention]**

**[0001]**

**[Technological Field of the Invention]** The present invention relates to a resin composition for calender molding comprising an amorphous polyester resin that is suitable for being molded into sheeting by means of calender molding.

**[0002]**

**[Prior Art]** Polyester resins such as polyethylene terephthalate (PET) have for many years generally been used extensively in fibers, films, and the like.

**[0003]** Among these applications, films designed for universal use have been transparent; exhibited exceptional tensile strength, impact strength, shear strength and other mechanical characteristics; and had a low temperature dependency in environments where normal working temperatures are encountered, with these attributes having been achieved by means of extruding the aforementioned resin in a molten state using extrusion-molding, rapidly cooling and causing the molten resin to solidify, stretching the resulting resin with a biaxial stretching technique to prompt crystallization, and subsequently causing the resulting article to set in response to subjecting the same to heating at a specific temperature. These films also have had exceptional electrical insulation properties and resistance to chemicals and solvents.

[0004] As has been described in the foregoing, a crystalline resin is used as a starting material in such films, and it is therefore essential to use stretching means in which a biaxial stretching technique or the like is employed in order to obtain a product having excellent mechanical characteristics and transparency. As a result, it has generally been impossible to obtain flat products ("sheeting" hereunder) that have a thickness of 0.3 mm or greater and can be referred to as sheeting.

[0005] Moreover, even if sheeting is obtained from the aforementioned resin, the fact that a crystalline resin is used causes low viscosity to be obtained during heating; prevents so-called fabrication as a result of thermoforming, which is performed with a technique such as vacuum- or pneumatic-molding; and creates complications during bonding with organic solvents or the like.

[0006]

**[Problems to Be Solved by the Invention]** In order to obtain sheeting from the aforescribed polyester resin that exhibits improved extrusion molding properties while also exhibiting the conventional mechanical characteristics and transparency within ranges that are permissible for implementation, means have been devised for improving the essential properties of the resin, with such enhanced resins enjoying widespread application in recent years. In other words, so-called amorphous polyethylene terephthalate, in which cyclohexane diol is substituted for a portion of the ethylene glycol when the dimethyl terephthalate and ethylene glycol are transesterified and polycondensed, is used as an extrusion-molding starting material when sheeting is to be obtained.

[0007] Nevertheless, a technical aspect that is well known in extrusion molding holds that the quantity of sheeting produced (i.e., discharged) per unit time is in general markedly less than that obtained using calender molding, due to the constraints inherent in the structure of the equipment used.

[0008] It has accordingly been believed that amorphous polyethylene terephthalate should be applied to calender molding in order to raise productivity; however, such resins have actually proven impossible to use alone in calender molding for the following reason. In other words, the resin exhibits low viscosity during thermoforming and has a high degree of adhesion on the surfaces of the metal calender rolls. Accordingly, the resin cannot be separated from the surfaces

of the metal rolls after having adhered thereto, and consequently sheeting has been impossible to form therefrom.

[0009] The present inventors perfected the present invention based on the discovery that as a result of selecting a specific lubricant, and adding the lubricant in a quantity within a fixed range with regard to the amorphous polyethylene terephthalate, which is used as a polyester resin, the resulting resin composition may be established as a starting material when thermoforming and fabricating sheeting using a calender molding technique.

[0010] In other words, it is an object of the present invention to provide a resin composition for calender molding comprising a polyester resin able to be molded into sheeting by means of calender molding.

[0011]

**[Means Used to Solve the Above-Mentioned Problems]** In order to achieve the aforementioned object, the main point of the present invention is a resin composition in which an amorphous polyester resin and a fatty acid ester-based montanic acid wax are included as essential components, wherein the resin composition for calender molding comprises a copolymer in which a portion of the ethylene glycol component among the terephthalic acid and ethylene glycol components of the amorphous polyester resin is substituted by 1,4-cyclohexane dimethanol.

[0012] According to Claim 2, a preferred embodiment of the present invention is the resin composition for calender molding according to Claim 1, wherein the blending ratio of the amorphous polyester resin and fatty acid ester-based montanic acid wax is 0.1 to 5.0 weight parts montanic acid wax per 100 weight parts of the amorphous polyester resin.

[0013] In the amorphous polyester resin, which is one of the essential components of the resin composition for calender molding of the present invention, a portion of the ethylene glycol component among the terephthalic acid and ethylene glycol component is substituted by 1,4-cyclohexanedimethanol, and the 1,4-cyclohexanedimethanol is used in a ratio of 10 to 70 mol% per 90 to 30 mol% of the ethylene glycol component.

[0014] If the ratio of 10 to 70 mol% 1,4-cyclohexanedimethanol per 90 to 30 mol% of the ethylene glycol component fails to be maintained, the crystallinity of the resin components will increase, and complications will be encountered when calendering is performed; accordingly, the

1,4-cyclohexanedimethanol is preferably used in a ratio of 20 to 60 mol% per 80 to 40 mol% of the ethylene glycol component.

[0015] Next, a fatty acid ester-based montanic acid wax, which is an ester compound of montanic acid and an alcohol, is applied as the lubricant used as the other essential component together with the amorphous polyester resin in the present invention. Examples of the alcohol include ethylene glycol, 1,2-butanediol, 1,3-butanediol, 2,3-butanediol, and glycerol. Partially saponified montanic acid waxes may also be used.

[0016] An appropriate amount of lubricant used with regard to the amorphous polyester resin is 0.1 to 5.0 weight parts per 100 weight parts amorphous polyester resin. If the amount is below 0.1 weight parts, severe adhesion will result between the resin composition and the surfaces of the metal calendering rolls, thereby preventing the resin composition from being separated from the metal rolls in the form of a sheet. If the amount exceeds 5.0 weight parts, less adhesion will result between the resin composition and the surfaces of the metal calendering rolls, but viscosity during melting will conversely decrease; therefore, the resin will be able to be removed from the surfaces of the metal rolls, but will elongate excessively under its own weight, which will ultimately prevent the resin from being formed into sheeting. Consequently, the amount of lubricant is preferably 0.5 to 2.0 weight parts per 100 weight parts amorphous polyester resin.

[0017] In addition to the amorphous polyester resin and montanic acid wax, which are essential components, a further lubricant may be added as a component in the resin composition for calender molding of the present invention as a release auxiliary for the metal calendering rolls. Examples thereof that may be jointly used include lubricants based on paraffin wax or other hydrocarbons; lubricants based on stearic acid or other fatty acids; lubricants based on stearyl amide or other fatty acid amides; and lubricants based on fatty acid esters, alcohols, metallic soaps, or the like.

[0018]

**[Embodiments of the Invention]** Embodiments of the resin composition for calender molding of the present invention shall be described below with reference to working and comparative examples. In the evaluations of moldability exhibited by sheeting obtained as a result of using a calendering apparatus in all of the working and comparative examples, a circle ("O") indicates a satisfactory result, and a cross ("X") indicates an unsatisfactory result. The ratios in the blends are indicated in terms of "weight parts."

**[0019] Working Examples 1 through 3**

A copolymer ("PET-1" hereunder) was prepared as the amorphous polyester resin. In this copolymer, a portion of the ethylene glycol component among the terephthalic acid and ethylene glycol components constituting the amorphous polyester resin had been substituted by 1,4-cyclohexanedimethanol, and the ratio of the components was 30 mol% of 1,4-cyclohexane dimethanol per 70 mol% of ethylene glycol.

[0020] An ester compound of montanic acid and an alcohol was prepared for use as the lubricant, and the montanic acid wax was prepared with 1,3-butanediol being used as the alcohol.

[0021] Next, a resin composition in which the amorphous polyester resin and montanic acid wax that had been prepared as described in the foregoing in the ratios shown in Table 1 was first heated and kneaded in advance as a pre-treatment using a twin-screw extruder, then conveyed to an L-shaped calendering apparatus comprising four metal rolls 250 mm in diameter, and calendered at roll temperatures of 170 to 180°C to mold 0.5 mm-thick sheeting.

[0022] According to Working Examples 1 through 3, the resin compositions exhibited no strong adhesion to the metal calendering rolls, had satisfactory release properties, and were able to yield sheeting of the anticipated thickness (0.5 mm).

[0023] Satisfactory results were obtained in all of the evaluations performed in regard to the ability of the working examples to be molded into sheeting, as is displayed in Table 1.

[0024]

[Table 1]

	Working Examples			Comparative Examples				
	1	2	3	1	2	3	4	5
(PET-1)	100	100	100		100	100	100	100
(PET-2)				100				
(Lubricants)								
Montanic acid wax	0.5	1.0	2.0	1.0	0.05	6.0		
Stearic acid							1.0	
Polyethylene wax								1.0
(Moldability)	○	○	○	×	×	×	×	×

**[0025] Comparative Examples 1 through 5**

In contrast to the aforementioned working examples, a crystalline polyester resin in which the ethylene glycol component had not been substituted by 1,4-cyclohexanedimethanol or

another compound; i.e., polyethylene terephthalate ("PET-2" hereunder), was used in Comparative Example 1 instead of the PET-1 used in the working examples; and amorphous polyester resins that were the same as the PET-1 used in the working examples were used in Comparative Examples 2 through 5. In Comparative Example 1, the same montanic acid wax used in the working examples was used as the lubricant, and in Comparative Examples 2 through 5, the lubricants used were selected from among the others cited in the foregoing as release auxiliaries for metal calendering rolls, and are also displayed in Table 1.

[0026] Next, the various resins and lubricants used in Comparative Examples 1 through 5 were blended in the ratios shown in Table 1, and attempts were subsequently made to use the calendering apparatus to mold sheeting having the same thickness, using the same procedure followed in the working examples.

[0027] The conditions during the molding procedure were such that in Comparative Example 1, the resin composition did not gel in the extruder at a cylinder temperature of 180°C, and could not be conveyed thereafter to the calendering apparatus. The resin did gel once the cylinder temperature had been elevated to 250°C, but the melt viscosity declined precipitously, and the extrudate adhered to the metal rolls after having been conveyed to the calendering apparatus, thereby preventing the article from being removed therefrom in the form of a sheet, which prevented a sheeting from being obtained. In Comparative Examples 2 through 5, all of the resin compositions gelled in the extruder at a cylinder temperature of 180°C, and were successfully conveyed to the calendering apparatus; however, the extrudates tightly adhered to the metal rolls, and could not be released smoothly therefrom, which prevented sheeting from being molded.

[0028] In other words, as shown in Table 1, the results of the evaluations of moldability were unsatisfactory for every type of sheeting obtained with the comparative examples.

[0029]

**[Effect of the Invention]** As has been described in the foregoing, the resin composition for calender molding of the present invention includes an amorphous polyester resin and fatty acid ester-based montanic acid wax as essential components, and comprises a copolymer in which a portion of the ethylene glycol component among the terephthalic acid and ethylene glycol components of the amorphous polyester resin is substituted by 1,4-cyclohexanedimethanol. Consequently, the viscosity of the resin composition during heating is appropriately high, and its

degree of adhesion to the surfaces of the metal calendering rolls is low, facilitating the separation of the resin from the rolls, enabling sheeting to be molded using calendering techniques, and allowing production per unit time to be dramatically improved.

[0030] Furthermore, according to Claim 2, the amorphous polyester resin and fatty acid ester-based montanic acid wax in the resin composition for calender molding according to Claim 1 are blended in a ratio of 0.1 to 5.0 weight parts montanic acid wax per 100 weight parts of the amorphous polyester resin, creating the advantage that the melt viscosity and degree of adhesion to the metal rolls may be suitably adjusted, and optimal calender molding conditions set, while the mechanical characteristics and transparency of the sheeting are retained within ranges that are permissible for implementation.